

Wetting of the Au(110) substrate: Homoepitaxial islands and layers

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The morphology of epitaxial structures formed in solidification of liquid Au films on the Au(110) surface was studied by molecular dynamics simulation based on a many-body interatomic potential. The (1×2) reconstructed and smooth phase at the temperature $T = 500$ K, as well as a deconstructed and rough phase of the Au(110) substrate at $T = 900$ K, were investigated. The three-dimensional islands with the (111) oriented facets were formed in solidification of thinner liquid films. At the same time, the substrate below these islands underwent a weak faceting process. Conversely, the solidification of thicker liquid films resulted in the flat solid films. The two surface phases of the Au(110) substrate induced different structure of these epitaxial solid films. The films were studied via construction of the Voronoi polyhedra, density profiles, and surface stress calculations.

I. INTRODUCTION

The wetting of substrates by liquid and solid adsorbates attracts interest in diverse fields. The connection of wetting and epitaxial growth is important for fundamental surface physics and at the same time for applications. The Au(110) surface has been investigated by many experimental techniques^{1,2}. In a temperature range from $T = 0$ K up to the bulk melting point $T = 1337$ K, the Au(110) surface exhibits several structural phases. The surface is at low temperatures smooth and reconstructed into the so-called “missing-row” (1×2) structure. In this phase the length of the surface unit cell is the same as in the bulk along $[1\bar{1}0]$, while it is twice as long along the $[001]$ direction. Alternating $[1\bar{1}0]$ rows are absent in the topmost layer. The missing row structure disappears in a phase transition of the Ising type at ≈ 650 K. In recent experiments the roughening transition on some well-characterized surfaces was studied³. At the roughening transition temperature the flat facet on equilibrium crystal shape becomes rounded and the step free energy of an interface goes to zero. For statistical mechanical models it is suitable to define the roughening transition where the height-height correlation function exhibits divergence. In particular, the Au(110) surface undergoes the Kosterlitz-Thouless roughening transition at ≈ 700 K^{2,3}. The Au(110) surface also exhibits surface melting: it starts to melt below the bulk melting temperature T_m and the thickness of the liquid layer diverges as T_m is approached⁴.

Barbier, Salanon, and Sprösser recently started investigation of the structure and ordering dynamics for the growth of Au on the (1×2) reconstructed Au(110) surface⁵. They have found that a metastable (1×1) phase is formed after deposition of a half of a monolayer at 190 K. The (1×2) disordered phase is formed after a deposition of one monolayer. The method of deposition of Barbier and coworkers included some features of the epitaxy from the liquid phase. They used a Knudsen type evaporator for liquid gold and obtained a flux of one monolayer in a few minutes. The presence of several surface phases on Au(110) calls for a detailed investigation of homoepitaxial growth on this substrate. Unfortunately it is often a formidable task to study solidification and growth at an atomistic level using now available experimental techniques. Computer simulations employing carefully chosen potentials for real materials may certainly help one to understand these phenomena. For example, results of MD simulations with the Stillinger-Weber potential are in good agreement with experimental observations of the silicon surface-melt interface after laser-induced melting^{6,7}. MD simulation study of a metal surface, in particular solidification of thin liquid Au films on Au(110), is presented in the following.

The structures formed in solidification of liquid Au films on the (1×2) reconstructed phase, as well as on a rough phase of the Au(110) surface are studied in this work. The MD simulation method is used to investigate the microscopic features of epitaxy. The results show that three-dimensional islands are formed at the substrate in solidification of thinner liquid films, whereas flat solid films grow in solidification of thicker films. Different structures of epitaxial films are found for a reconstructed and rough substrate. The structure of the films was investigated via monitoring of real-space particle trajectories, density profiles, surface stress values, and construction of the Voronoi polyhedra. In the following the MD technique, and the potential and simulation methods are described in Sec. II. Results are presented in Sections III–V. A discussion and the conclusions are given in Sec. VI.

II. SIMULATION MODEL AND METHOD

MD simulations were used to study epitaxial solidification of liquid Au films on Au(110). The interatomic interactions were derived from a many-body classical Hamiltonian^{8–10}. This kind of potentials gives a good characterization of interactions and cohesion in metals⁹. All details of the so-called “glue” model potential were

fixed before and there were no arbitrary parameters in the present simulation¹⁰. This potential is given by

$$V = \frac{1}{2} \sum_{\substack{i,j=1 \\ (j \neq i)}}^N \phi(r_{i,j}) + \sum_{i=1}^N U(n_i), \quad (1)$$

where the density n_i is a superposition of two-body contributions

$$n_i = \sum_{\substack{j=1 \\ (j \neq i)}}^N \rho(r_{i,j}). \quad (2)$$

In the glue model all three functions $\phi(r)$, $U(n)$ and $\rho(r)$ were included in the fitting procedure¹⁰. The glue potential for gold is well tested and known to reproduce experimental results for many diverse bulk, surface and cluster properties. Garofalo, Ercolessi, and Tosatti used this potential to study the reconstruction on the Au(110) surface¹¹. They proved that the (1×2) missing row structure has the lowest surface energy. Bernasconi and Tosatti studied the Au(110) surface structure within the glue model at temperatures around deconstruction and roughening². These transitions in MD simulation are characterized with slow kinetics. In addition, a large lateral size of MD box is necessary due to the logarithmic divergence of the correlation function at the roughening transition. Therefore, Bernasconi and Tosatti showed that it is not feasible nowadays to study the thermodynamical characteristics of the roughening transition on Au(110) using MD method.

The simulation started from the model for the substrate, but with a goal of modeling the epitaxy on Au(110). Separate MD slabs were used to study the growth on a reconstructed and rough surface. The temperature was controlled by rescaling the particle velocities. The lattice constants of all samples were changed with temperature according to the expansion coefficient of the bulk. This coefficient and the bulk melting temperature ($T_m \approx 1355$ K) were known from the previous bulk simulation¹⁰. The melting point in the glue model is in good agreement with the experimental value of 1337 K. MD boxes consisted of 12 layers and were constructed to have the (1×2) reconstructed top layer at $T = 0$ K. The three bottom layers were kept fixed to model the bulk. The number of atoms was 10 along the $[001]$ direction and 5 along $[1\bar{1}0]$. These periodically repeated MD slabs for the substrate were made up of 575 particles. The simulations for MD boxes with 10 and 20 atoms along the $[1\bar{1}0]$ direction (consisting of 750 and 1500 particles, respectively) were also performed. No size dependence was observed, and only the detailed results for the smallest MD box will be presented here. As usual, for bigger samples and larger phase space longer MD simulations are needed to achieve the same structure. This requirement is crucial for MD studies of thin film growth. The samples were heated up to $T = 500$ K = $0.37T_m$ for a

reconstructed phase, and $T = 900$ K = $0.66T_m$ for a rough phase. At these temperatures runs of 5×10^5 MD time steps (3.6 ns) for a smooth phase and 10^6 MD steps (7.1 ns) for a rough phase were done. Bernasconi and Tosatti found that a long equilibration is necessary to obtain the rough phase on Au(110)². The final configurations of MD boxes representing the Au(110) substrate are shown in Fig. 1. Two and four additional solid layers were then deposited on the top of MD boxes to follow the stacking of the Au(110) surface. In this paper these new epitaxial atoms are referred to as thinner films (two initial solid layers) and thicker films (four initial solid layers). The structures of the Au(110) substrate were preserved by temporarily fixing the corresponding particles. The epitaxial films were warmed up to $T = 2000$ K = $1.48T_m$, and then equilibrated for 0.36 ns. No atom was observed to evaporate from the slab surface. This agrees with the experimental evaporation rate of gold which is in the range from $50 \text{ atoms s}^{-1} \text{ \AA}^{-2}$ at 1360 K to $6 \times 10^4 \text{ atoms s}^{-1} \text{ \AA}^{-2}$ at 3000 K¹². The samples with liquid film on the top were cooled back to 500 K and 900 K and the particles representing the substrate were released. The time evolution of substrates and epitaxial films was then followed for additional 3.6 ns. Some simulations for deposition and solidification were repeated for the second MD box of the same size and similar results for the morphology of epitaxial structures were obtained.

III. MORPHOLOGY

Figure 2(a) shows the result of solidification of a thinner liquid film on the smooth, (1×2) reconstructed substrate after 5×10^5 time steps. The faceted structure of the solid film is evident. The big island consists of two connected four-layer high islands. The facets are with the (111) structure and they are oriented under the angle of 35° to the substrate. A small island on the left was also formed. Figure 2(b) shows the result of solidification of a thinner liquid film on the rough Au(110) surface after 3.6 ns of simulation. One four-layer high island with a flat top was formed. The facets on the island are disordered, with the (111) structure and oriented under the angle of 35° to the substrate. Underlying substrate exhibits faceting: four substrate atoms (two on each side of the box) moved to the island and as a result longer (111) facets were formed. The (1×2) reconstructed substrate shown in Fig. 2(a) also tends to facet after solidification of a liquid film.

Figure 3 represents the structure formed in solidification of a thicker liquid film on the smooth, (1×2) reconstructed Au(110) surface after 3.6 ns of simulation. The film is flat and its surface is disordered. Deposited atoms first filled the missing rows of the substrate and then more disordered layers were formed on the top of the film. The observed trends agree with the results of Barbier *et al*⁵. The structure formed in solidification of

thicker liquid film on a deconstructed, rough substrate after 3.6 ns of simulation is shown in Fig. 4. The film is flat and more ordered along the $[100]$ direction than along $[1\bar{1}0]$. The side view in Fig. 4(a) shows that stacking faults exist at the rough substrate–film interface. The top layers of the rough substrate are more ordered after than before deposition. This is the case for both thinner and thicker films [shown in Figs. 2(b) and 4, respectively].

IV. STRUCTURE

The reason for the island formation in deposition of thinner films was a tendency of gold atoms to increase their coordination. It is well known that a small coordination of atoms at gold surfaces provides a driving force for their reconstruction, i.e., the formation of denser layers with a higher coordination. It was calculated that the average coordination was 6.044 neighbors for the initial, ideal solid configuration of a thinner film. The average coordination increased to 9.068 neighbors for the initial solid configuration of a thicker film. The small coordination of ≈ 6 caused the clustering, i.e., the change of the structure that increased the coordination number. The three-dimensional islands, therefore, were formed in solidification of thinner films. The higher coordination of atoms in thicker epitaxial films was of the same order as for reconstructed gold surfaces and the flat films were formed.

The densities projected on the vertical axis for the flat epitaxial films are shown in Fig. 5. The films are less ordered than the substrate, but the layered structure exists on the density plots.

Construction of the Voronoi polyhedra was done to describe an amorphous structure of the flat solid films¹³. The Voronoi polyhedron is a topological generalization of the Wigner–Seitz cell for a crystal. It is defined for a given atom as the region consisting of all points nearer to it than to any other atom. The Voronoi polyhedra characterize the local atomic configurations and disorder for a given phase. Figure 6 shows the distribution of the number of neighbors for the Voronoi polyhedra constructed for the atoms in the flat solid films. The coordinates of particles are taken from MD simulations after the time evolution of 3.6 ns, i.e., for the structures shown in Figs. 3 and 4. Only the particles in the films were analyzed, but the atoms in the first layer of the substrate were also considered as neighbors. The particles were not assumed to be neighbors beyond the cutoff of 5 Å. The average number of the Voronoi neighbors for a thicker film on a smooth substrate is $\langle i \rangle = \sum_i i n_i = 14.19$, whereas for a thicker film on a rough substrate $\langle i \rangle$ is 14.96. These values agree with other reported results for the number of faces of the Voronoi polyhedra in three-dimensional systems¹³. The average value of this number is 6 in two dimensions, whereas it is 14 in three dimensions and for regular close packing. Using MD simulation

Rahman found 15.67 for the randomly distributed atoms of ideal gas, 14.45 for the liquid argon and 14.26 for the solid argon¹³. Finney found the value of 14.2 for a random close packing of hard spheres using the Monte Carlo simulation¹³. Figure 6 shows that the maximum of distribution for the number of faces of the Voronoi polyhedra corresponds to $i = 13$ for the film on a smooth substrate, and to 12 for the film on a rough substrate. The distributions are broad and represent a disorder, i.e., the presence of topologically defective polygons where the number of faces is not 14.

V. SURFACE STRESS

When the film is deposited on the surface at certain temperature there is a competition between the requirements of bonding to the substrate and a tendency of particles to adopt the minimum energy separations in the epitaxial layers. These interactions are origin of the surface stress in the film. The surface of the substrate is by itself in a stressed state because of different bonding than in the bulk. The surface stress tensor $\sigma_{\alpha\beta}$ is given by

$$\sigma_{\alpha\beta} = \gamma \delta_{\alpha\beta} + \frac{\partial \gamma}{\partial \epsilon_{\alpha\beta}}, \quad (3)$$

where γ is the surface tension, $\delta_{\alpha\beta}$ is the Kronecker symbol, $\epsilon_{\alpha\beta}$ is the surface strain tensor, and α, β are directions in the surface plane¹⁴. In MD simulation the surface stress can be obtained from the components of the pressure tensor¹⁵. In computation of the stress MD slabs for the substrates and thicker films described in Sec. II were used. MD box representing the (1×2) reconstructed Au(110) surface at $T = 0$ K was equilibrated for 0.71 ns. The values of surface stress along the $[1\bar{1}0]$ and $[001]$ directions that describe the anisotropy of the (1×2) reconstructed Au(110) substrate are presented in Table I. The components of the surface stress tensor along and perpendicular to the close packed rows of a reconstructed substrate are different even for the films. The stress is smaller along the close-packed rows for a reconstructed Au(110) surface at $T = 0$ K. As the temperature increases this value becomes bigger than the other component, for both the substrates and films. The same relationship was obtained by Toh, Ong and Ercolessi for the Pb(110) surface¹⁶. Table I also shows that the stress for the (1×2) Au(110) surface at 500 K decreases after deposition of the film. On the contrary, the surface stress increases when the film is deposited on the rough substrate at 900 K. In general, the surface stress values found in these simulations are similar to those found in other recently reported calculations for the (110) surfaces in fcc metals^{16–20}. The surface stress can be in principle calculated by ab-initio electronic structure calculation and MD simulation. For all (110) surfaces the stress is positive, i.e., tensile and favors contraction of the lattice. The

experimental data for the surface stress on well-defined metallic surfaces do not exist. The surface stress value of $0.073 \text{ eV}\text{\AA}^{-2}$ was obtained at 323 K in measurements for the radial strain in small Au spheres¹⁴. Solliard and Flueli found (at 300 K) the value $0.1922 \text{ eV}\text{\AA}^{-2}$ for the (220) ring on a small gold sphere, and $0.1991 \text{ eV}\text{\AA}^{-2}$ for the (422) ring²¹. For comparison, the average values of the surface stress tensor for the substrates and films studied in this work are also given in Table I. More studies, and especially measurements, for the stress on metal surfaces and films are necessary. The change in the surface stress after deposition of a layer of Ga on the Si crystal was measured²².

VI. DISCUSSION AND CONCLUSIONS

The results of simulation show solidification of liquid Au films on the Au(110) surface at 500 K and 900 K. This problem is equivalent to the wetting of the Au(110) substrate by thin films of gold²³. The morphological change of the film with its thickness is a transition from nonwetting to wetting behavior. The initial deposited solid films were warmed up to $T = 2000 \text{ K}$. Therefore undercoolings of 1500 K for a smooth phase and 1100 K for a rough phase of the substrate were used in the simulations. At these large undercoolings the growth and solidification can be viewed as continuous processes, i.e., a nucleation rate is high²⁴. As a consequence, at both temperatures solidification may proceed from any point on the surface and there is no requirement for the special nucleus as in a nucleation mechanism. Therefore in this simulation no difference in the growth mode between a smooth and rough substrate was found. This is consistent with a view that in solidification smooth surfaces undergo a kinetic roughening transition already for small driving forces²⁴. As a result only rough surfaces are effectively present in solidification. The (1×2) reconstruction on the smooth substrate was naturally removed because the deposited atoms first filled the missing rows.

The facets on the three-dimensional islands formed in solidification of thinner films were (111) oriented. The substrate below these islands exhibited faceting. The small facets of the substrate joined the big (111) facets of the islands. The (111) facets are also formed in faceting of gold vicinals induced by surface reconstruction and surface melting²⁵. The (111) face of fcc metals has the lowest surface energy and the structures develop to “open” the (111) orientation. Recently faceting induced by deposition of metallic films was found for the bcc substrates, such as W(111) and Mo(111)²⁶. For example, a monolayer of Pd on W(111) causes the formation of the (211) oriented facets. Although faceting is already in the focus of the current interests in surface physics, this phenomenon certainly needs further investigation²⁷. This is especially true for faceting of the substrates induced by deposition of thin films.

The method of the film preparation used in this work resembles the high-temperature annealing in simulations for a structural investigation of free clusters. The techniques of thermal annealing and rapid thermal annealing are also used in experiments for crystallization of amorphous thin films. The direct high-temperature annealing procedure gives (within reasonable MD simulation time) amorphous structures of low energies for clusters bigger than 60 particles²⁸. In this simulation the substrate tends to order the particles during solidification. Similar method of the high-temperature surface annealing of free Au clusters gives better results than other techniques²⁸. In this method only the particles in the external shell are allowed to move, while the internal particles of the cluster are kept frozen in the crystalline structure. The method of the film preparation at the high temperature, used in this work, simulates the deposition from the liquid phase and should be important for MD studies of epitaxy. It is well known that in MD simulations of the film growth from the vapor phase a big problem is insufficient computer time to follow experimental deposition rates. MD simulations done with extremely high deposition rates sometimes produce unphysical effects. The simulations where the solid films of a certain thickness are directly placed on the substrate are far from the experimental reality. In addition, if such a method is used at low temperatures and for thick films, then it is difficult to achieve real equilibrium structure in a limited simulation time. The method of the high-temperature annealing of thin films gives different solution for MD simulation of epitaxy.

In summary, solidification of liquid Au films on the Au(110) substrate was studied. MD simulation method, based on a many-body interatomic potential of proven accuracy, was used. The results showed that three-dimensional islands were formed in solidification of thinner liquid films. The substrate below these islands exhibited faceting. Flat solid films grew in homoepitaxial solidification of thicker liquid films. The films deposited on the smooth, (1×2) reconstructed phase at $T = 500 \text{ K}$ and on the rough, deconstructed phase of the substrate at $T = 900 \text{ K}$ were studied. Epitaxial solid films of different structure were formed on these substrates. Surface phases of Au(110) did not change the wetting mode of homoepitaxial films. Instead, the thickness of the film determined homoepitaxial growth mode on the Au(110) substrate. The detailed experimental studies of homoepitaxial growth on the Au(110) substrate, at all temperatures and for a various thicknesses of the film, are necessary.

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FIG. 1. Atomic configurations of molecular dynamics slabs used as a substrate. Two boxes along the $[1\bar{1}0]$ direction are shown. (a) The (1×2) reconstructed and smooth surface: a missing row structure is present at 500 K. (b) A deconstructed and rough surface at 900 K: the missing row structure of the substrate disappears.

FIG. 2. Particle trajectories showing the side view of molecular dynamics boxes after an equilibration time of 3.6 ns for a thinner film: (a) on the (1×2) reconstructed Au(110) surface at 500 K, (b) on a rough Au(110) surface at 900 K. All trajectory plots refer to a time span of 7 ps and show only the moving particles. For the side views trajectories display only nine substrate layers and the five $[1\bar{1}0]$ planes overlapped. The arrows point to the top substrate layer. Note faceting of the substrate in Fig. 2(b): four atoms in the two top layers of the substrate are missing, remaining microfacets are (111) oriented and join the facets of the island.

FIG. 3. Thicker film on the (1×2) reconstructed Au(110) substrate at 500 K. (a) Side view of the box. Deposited particles first fill the missing rows in the top (i.e., ninth on this figure) layer of the substrate, the next layer of the film is regular and follows the stacking of the substrate. The top region of the film is amorphous. (b) Top view of the film.

FIG. 4. Thicker film on a rough Au(110) surface at 900 K. (a) Side view of the box along the $[100]$ direction: the top (i.e., ninth on this figure) layer of the substrate is more ordered below the film than before deposition [corresponding rough substrate is shown in Fig. 1(b)]. (b) Top view of the film. (c) Side view of the box along the $[1\bar{1}0]$ direction.

FIG. 5. The (x, y) averaged density profile for molecular dynamics box simulating solid films grown from thicker liquid films. The three leftmost layers of the box are kept fixed and the film is on the right. (a) Film on a reconstructed substrate at 500 K. (b) Film on a rough substrate at 900K.

FIG. 6. The fractional concentrations for the number of neighbors, i.e., faces of the Voronoi polyhedra. (a) Thicker film on the (1×2) reconstructed Au(110) substrate at 500 K. (b) Thicker film on a rough Au(110) substrate at 900 K. The numbers of neighbors not equal 14 represent the extent of amorphousness. The thick vertical lines show the numbers, whereas the thin line is the envelope.

TABLE I. Surface stress $\sigma_{\alpha\beta}$. The components of the stress along and perpendicular to the close packed rows of the (1×2) reconstructed Au(110) surface, as well as their average value are shown. All values are in $\text{eV}\text{\AA}^{-2}$.

Surface structure	$[\bar{1}\bar{1}0]$	$[001]$	Average
Film on the smooth substrate, 500 K	0.101	0.063	0.082
Film on the rough substrate, 900 K	0.155	0.124	0.140
(1×2) Au(110), 500 K	0.199	0.148	0.174
Rough Au(110), 900 K	0.130	0.118	0.124
(1×2) Au(110), relaxed, 0 K	0.184	0.191	0.188

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